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16. ABSTRACT

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Size of Airborne Particulate Matter Over Three-Year Period

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JOURNAL ARTICLE

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■ A cascade impactor network operating from 1970 through 1972 at eight urban and two nonurban sites indicated that the median size of airborne particles on a mass basis appeared to increase over the sampling period. Quarterly variations in the median size were associated with seasonal changes, decreasing in summer but increasing in winter. The general upward trend in the size of airborne particles appeared to be associated with the reduction in automotive emissions that can lead to the formation of fine, secondary aerosols. Other possible explanations, however, were examined in addition to an evaluation of the limitations of the cascade impactor used in the network.

Recent investigations have revealed that the concentration of chemical components in airborne particulate matter can vary markedly with particle size (*1–5*). Characterizing the size of airborne particulate matter and its chemical composition is important to understand their effects on health and atmospheric properties and to identify their sources and environmental sinks.

The National Air Surveillance Cascade Impactor Network was established in 1970 in an effort to determine the aerodynamic size of airborne particulate matter on a mass basis at urban and nonurban sites and to assess changes in the size of particulate matter over a long time period (*6*). Measurements at six cities indicated that suspended particles were predominantly submicrometer in diameter with a mass size distribution that appeared to be reasonably well described by a single mode log normal function. These results were in good agreement with available measurements made before 1970 with other types of aerodynamic separators. The relationship of selected chemical constituents to the particle size of airborne particulate matter was also determined for the 1970 network operation (*2*).

The network was expanded in 1971 to a total of 10 stations with the addition of two urban sites, Seattle and Steubenville, and two nonurban sites, Grand Canyon and Manteo. Although complete data over the full three-year period were available for only a few stations, sufficient data were collected at most of the urban sites to reasonably characterize long-term variations in the size and concentration of airborne particulate matter. In addition, an extensive evaluation of the modified Andersen impactor was made including verification of the theoretical calibration with laboratory-generated aerosols.

Experimental

The modified Andersen cascade impactor used in the network and the sampling and analytical methodology have been described previously (*2, 6*). Briefly, however, a six-stage Andersen impactor (*7*) designed to operate at 1 cfm (0.028 m^3/min) was modified by removal of the last stage so that the operating air flow rate could be increased 3–5 cfm (0.085–0.142 m^3/min) to collect a sufficient quantity of material for gravimetric analysis. Particulate matter was impacted on uncoated aluminum foil collection surfaces placed in each stage while a backup filter was used to collect unimpacted particles. After a 24-h sampling period, the collection surfaces were returned to the laboratory for gravimetric analysis and subsequent calculation of the particle size distribution.

There are a number of distinct advantages in using a modified Andersen impactor for determining the size of airborne particulate matter. Sizing particulate matter aerodynamically simulates the action of the respiratory system, thereby providing a direct estimate of particle deposition and retention in the lungs. Because particulate matter is physically collected in sufficient quantities for gravimetric and chemical analysis, the mass size distribution can be determined directly, expressed as spheres of unit density. In addition, the impactor is rugged, reasonably low in cost, and simple to operate—features that are attractive for field monitoring projects.

On the other hand, the modified Andersen impactor, as all measurement instruments, is beset with inaccuracies and limitations which can affect the usefulness of resulting data. The limitations of the sampler have been described previously (*8–11*); however, laboratory and field evaluations have since been made and are reported below.

Results obtained with the modified Andersen impactor generally agree quite well with results from other aerodynamic size fractionating samplers. Mass median diameter values for suspended particulate matter determined with the Lundgren impactor (*5, 12–14*) and the median size of chemical components reported by other investigators (*1, 4, 5, 15–18*) exhibit a remarkable consistency with our previously published results (*2, 3, 6, 19*). Direct side-by-side comparisons of the modified Andersen, Lundgren, and Scientific Advances impactors carried out by Gordon et al. (*20*) indicated that the Scientific Advances and modified Andersen impactors were subject to particle bounce-off and wall loss errors estimated to be up to 20%.

Calibration. Calibration of the modified Andersen impactor was derived from the theory of Mercer (*21*) and Rans and Wong (*22*) as described previously (*6*). The theoretical calibration expression relates the collection efficiency for each stage as a function of size to the average air flow rate during sampling which ranged from about 3 to 5 cfm (0.085–0.142 m^3/min). Relying on a calibration with laboratory-generated aerosols has several drawbacks which include the fact that laboratory test aerosols are unlike airborne particulate matter, are extremely difficult and time consuming to generate, and can suffer from inherent inaccuracies in use (*23, 24*).

The Andersen impactor and its modifications have been calibrated at 1 cfm (0.028 m^3/min), 3 cfm (0.085 m^3/min), and 5 cfm (0.142 m^3/min) using various test aerosols including latex spheres, dioctylphthalate (DOP), and dye solutions and various methods of aerosol generation. A comparison of laboratory calibration results with the theoretical prediction is summarized in Table I. A comparison of laboratory results at the 1 cfm flow rate, which would be easiest and most accurate to carry out, shows some variability but is in reasonable agreement with the theoretical prediction.

Calibration at 3 and 5 cfm yielded greater divergence from the theoretical prediction on the lower stages, although quite good agreement was found on the upper stages. Burton's (*20*) calibrations using the Berglund and Liu (*30*) aerosol generator are most meaningful, however, because the impactor was operated under field conditions, i.e., with aluminum collection plates and rainshield in place. Although there is reasonably good agreement with the calibration results on the upper stages, the Burton calibration appears to indicate poor particle size separation characteristics for the lower stages, especially

Table I. Calibration of Andersen Sampler—Comparison of Theoretically Derived D_{50} Values with Laboratory-Determined Calibrations (Expressed in μm)

Stage	Thiel (31)	Andersen 1/1	May (28)	Fitzsh et al. (30)	Yeas et al. (27)
		1 cfm (0.028 m³/min)			
1	7.0	9.0	N/A	N/A	12–30
2	4.9	5.5	5.5	5.4	5.5–7.0
3	3.3	3.3	3.5	3.0	N/A
4	2.1	2.0	2.0	1.8	1.9
5	1.1	1.0	1.1	0.9	0.8
		2 cfm (0.056 m³/min)			
		Nu (29)	Burton (29)		
1	5.1	4.0	4.2		
2	2.8	2.5	2.0		
3	1.9	1.5	1.0		
4	1.2	0.9	<0.6		
5	0.6	0.4	<0.6		
		5 cfm (0.142 m³/min)			
			Burton (29)		
1	3.2		3.1		
2	2.1		1.1		
3	1.4		<0.6		
4	0.9		<0.6		
5	0.4		<0.6		

at the 5-cfm flow rate. However, because of the possibility that the Berglund and Liu monodispersed aerosol generator may not run constant in the finer particle mode (*31*), the results of the Burton calibration for the lower stages should be regarded as suggestive at this time.

On the other hand, Burton's studies showed that particle bounce-off was negligible with the DOP aerosol; however, it is likely that solid crystalline material would be subject to greater bounce-off effects as observed by Gordon et al. (*30*) than would liquid DOP test droplets.

Table I indicates that the theoretical calibration expression for the modified Andersen impactor is reasonable for the upper stages but may tend to overstate the particle size for the lower stages, probably because of wall loss effects with the smaller particles.

The modified Andersen impactor appears to provide a reasonable estimate of the median size of airborne particulate matter on a mass basis. However, the authors do not think that the impactor possesses sufficient resolution to clearly define multimodal characteristics in the particle size distribution although recalculations of our data by other investigators (*11, 32*) suggest the distribution may be bimodal. Previous expressions of the size distribution as single mode log normal were somewhat of an oversimplification, especially in view of the recent calculations made by Durham et al. (*32*) which showed that impactors tend to exhibit poor size classification intervals on the lower stages which "wash out" the size resolution for smaller particles in the distribution. In the work reported here, the distributions are plotted on log probability paper which may obscure the multimodal characteristics of the distributions, if present, but provides quantitative information on inhalation indices such as mass median diameter and the percent respirable fraction. The particle size data from the modified Andersen impactor, however, can be especially useful for trend analysis, i.e., how the relative size distribution changes with time and location.

Network Operation. The cascade impactor network operated from 1970 to 1972 at six urban stations: Chicago, Cincinnati, Philadelphia, Denver, Washington, D.C., and St. Louis. Reasonably complete data for the three-year period

Table II. Average Annual Particle Size Distribution Parameters for 1970, 1971, and 1972

	1970	1971	1972
Chicago			
No. samples	51	55	59
Av concn, μg/cm³	66.5	55.6	57.9
Av MMD, μm	0.63	0.75	1.10
Av geo SD	6.2	5.5	5.9
Cincinnati			
No. samples	18	25	25
Av concn, μg/m³	74.3	57.5	47.6
Av MMD, μm	0.54	1.11	1.36
Av. geo SD	5.5	5.0	5.7
Philadelphia			
No. samples	20	15	
Av concn, μg/m³	58.5	49.5	
Av MMD, μm	0.47*	1.33	
Av geo SD	5.7	5.67	
Denver			
No. samples	21	17	12
Av concn, μg/m³	59.7	67.0	54.0
Av MMD, μm	0.40*	1.50	1.06
Av geo SD	10.5	11.1	7.2
Washington, D.C.			
No. samples	23		
Av concn, μg/m³	56.3		
Av MMD, μm	0.46*		
Av geo SD	5.2		
St. Louis			
No. samples	22	24	24
Av concn, μg/m³	73.1	61.1	64.5
Av MMD, μm	0.69	0.96	1.46
Av geo SD	6.8	6.1	5.0
Steubenville			
No. samples		23	12
Av concn, μg/m³		74.0	101.5
Av MMD, μm		1.09	1.52
Av geo SD		5.3	3.1
Seattle			
No. samples		22	18
Av concn, μg/m³		35.3	50.3
Av MMD, μm		0.52	1.25
Av geo SD		7.4	3.6
Grand Canyon			
No. samples		11	
Av concn, μg/m³		47.0	
Av MMD, μm		1.62	
Av geo SD		8.2	
Manteo			
No. samples		18	
Av concn, μg/m³		48.6	
Av MMD, μm		1.58	
Av geo SD		7.2	

*Value obtained from a short extrapolation of the distribution curve (*31*).

were obtained for Chicago, Cincinnati, Denver, and St. Louis; data over a two-year period were obtained for Philadelphia, and data over one year were obtained for Washington, D.C. In 1971 two additional urban stations were established at Seattle, Washington, and Steubenville, Ohio, and operated through 1972. Nonurban stations were operated at Grand Canyon, Arizona, and Manteo, N.C., in 1971. Sampler malfunctions precluded the collection of complete data for more than one year at the Grand Canyon and Manteo sites. Size distribution curves were calculated from the theoretical relationships described previously (*31*); however, the distributions were displayed graphically as log normal curves without

attempting to develop a least squares or other relationship which could closure possible multimodal characterization as in the previous report (6) contributing to some slight changes in the distribution parameters.

Results

Table II presents a summary of the average annual particle size parameters for the network operation, 1970 through 1972. In 1971 and 1972, Steubenville exhibited the highest annual average concentrations of 74.0 and 101.5 $\mu g/m^3$ respectively; for the same period, Seattle exhibited the lowest concentrations of 35.5 and 36.5 $\mu g/m^3$, respectively—levels that were below those found at the nonurban sites. Except for Steubenville and Denver, the average annual airborne particulate concentrations declined progressively over the sampling period.

Figures 1 and 2 present composite average annual particle size distributions for 1970, 1971, and 1972. Except for Denver, the annual average mass median diameter (MMD) for the six original urban sites (Figure 1 and Table II) increased progressively with a range of 0.40–0.69 μm in 1970, 0.75–1.32 μm in 1971, and 1.06–1.46 μm in 1972. Steubenville and Seattle, added in 1971, also exhibited an increase in the annual average MMD as seen in Figure 2. In 1971 the largest average MMD values were found at the nonurban sites, 1.69 μm in Manteo and 1.82 μm at the Grand Canyon; the smallest MMD for that year was found at Seattle with 0.59 μm. At sites where data exist for more than one year, a progressive decrease in the average geometric standard deviation was found, suggesting a narrower distribution or range of particle sizes with time.

Although the annual average MMD appeared to increase as a function of time, a more meaningful assessment can be seen in Figure 3 where the quarterly average MMD is plotted for each site. At Cincinnati, St. Louis, and Chicago where data are most complete over the three-year period, a general trend toward an increasing MMD can be seen despite quarterly variations. Similar trends can be seen at the other urban sites, although more intense variations in size are apparent. It is especially noteworthy that the variations in the quarterly MMD appear to follow a similar pattern at many of the sites, i.e., the average MMD decreased the second and third quarter but increased during the fourth and first quarter.

In light of the data on concentration, median diameter, and standard geometric deviation, a general pattern of the changing trend of airborne particulate matter on a weight basis can be seen at most urban sites. Over the three-year sampling period, the concentration of particulate matter at the urban sites has gradually decreased with the particle size distribution becoming narrower and reflecting a larger median size. On the other hand, the two nonurban sites generally exhibited particulate matter of a larger average median diameter, probably reflecting the influence of windborne dust (Grand Canyon) or sand and sea spray (Manteo).

Discussion

The generally accepted concept of airborne particulate matter attributes most of the large or coarse particles, about 3 μm diameter and above, to natural processes such as wind erosion, sea spray, and reentrained dust. These coarse primary particles are predominantly water insoluble and make up the major portion of the aerosol mass, perhaps up to 90% at some sites. Sedimentation controls the upper limit of the size spectrum although impaction on surfaces and wet precipitation (wash out) also remove the coarse particles. This concept is borne out in large measure by the particle size measurements at the Grand Canyon and Manteo sites where airborne

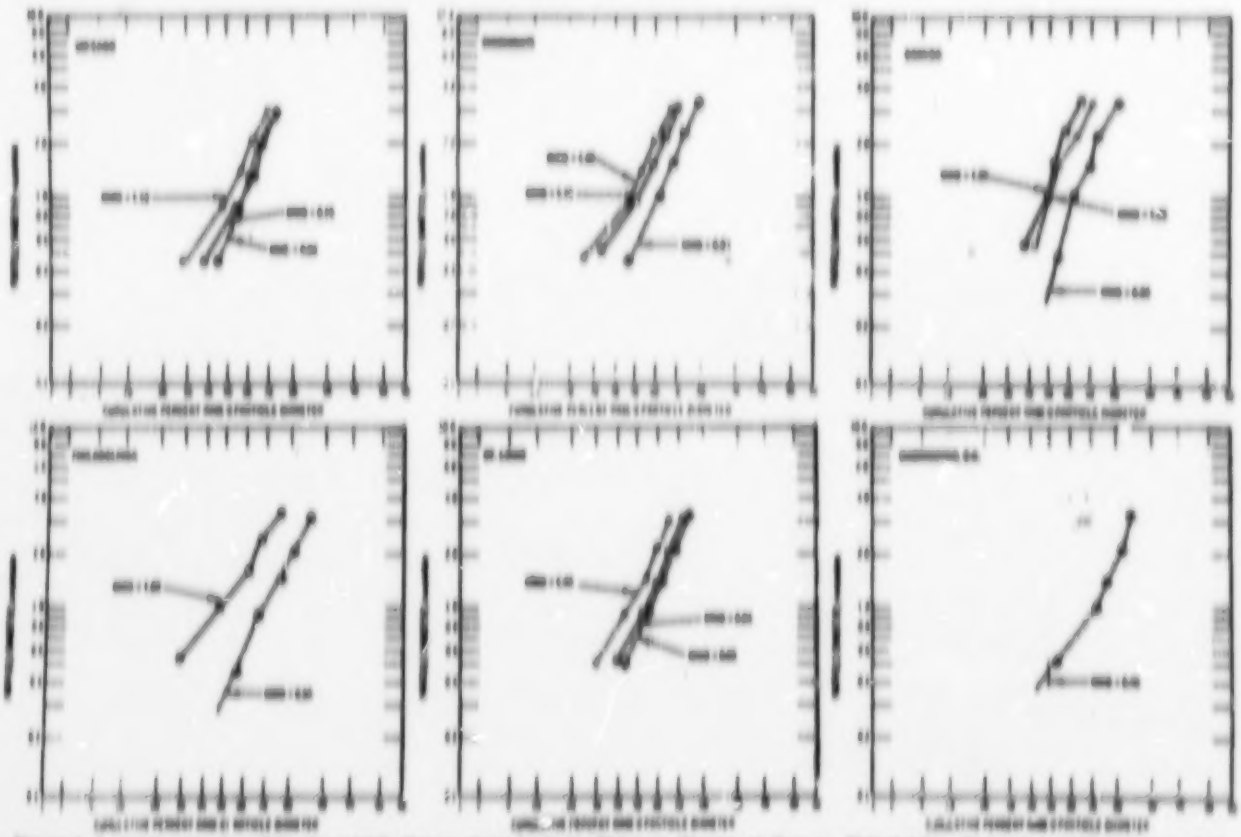

Figure 1. Average annual particle size distribution on mass basis for Chicago, Cincinnati, Denver, Philadelphia, St. Louis, and Washington, D.C., for 1970 (● · · · ●), 1971 (○ — ○), and 1972 (■ · · · ■)

dust and sand constitute the major portion of aerosol. Table II indicates the average annual MMD's for the Grand Canyon and Manteo were 1.63 and 1.58 μm, respectively.

Particles which constitute the lower end of the aerosol size spectrum, about 3 μm diameter and below, are generally associated with anthropogenic sources. These fine particles mostly consist of secondary reaction products, such as result from the conversion of gaseous pollutants to aerosols, and are composed in large measure of water soluble, hygroscopic material. In urban areas the major portion of the fine particles is composed of sulfate, nitrate, ammonium, lead, and hydrocarbon-containing material. The lower end of the size spectrum is controlled by agglomeration of gaseous molecules to form solid or liquid aerosols and by coagulation of particles which form larger particles by collision through Brownian motion. Coagulation, therefore, continually removes the fine airborne particles and enhances the number of coarse particles.

Whitby and coworkers (*34, 35*) have found evidence of the multimodal nature of atmospheric size distributions based on particle count measurements. Under photochemical smog conditions, the calculated volume distribution was almost always bimodal with a minimum at about 1–2 μm diameter, providing strong support for the concept of anthropogenic vs. natural sources of airborne particles. Unfortunately, cascade impactors generally do not provide sufficient resolution of the size spectrum to define multimodal characteristics of the distribution on a mass basis, especially at the lower end of the spectrum as discussed in the Experimental section.

The most significant finding in the study reported here is the apparent increase in the average median size at most urban sites over the three-year sampling period. The observed reduction in the average mass concentration from 1970 through 1972 is in agreement with the national trend determined by the Hi-Vol sampling network (*36*). The decrease in the median mass concentration has been attributed to the effectiveness of air pollution control measures which have also resulted in the reduction of gaseous sulfur dioxide concentrations nationally (*36*). It is not easy, however, to explain the corresponding increase in the median size of airborne particulate at most urban sites. Furthermore, although other workers have reported mass size distribution measurements at various sampling sites in the past, there is an insufficient amount of long-term trend data with which to compare our results.

Although the overall trend in the size of the MMD appears to be increasing with time, the quarterly variations seem to be related to season. From October through March, i.e., the fourth and first quarter, the average MMD appears to increase at all sites almost without exception as seen in Figure 3. It is likely that increased heating activities which can emit greater quantities of large airborne particles from ash and incomplete combustion and a reduction in photochemical activities which produce fine secondary particles account for this trend. On

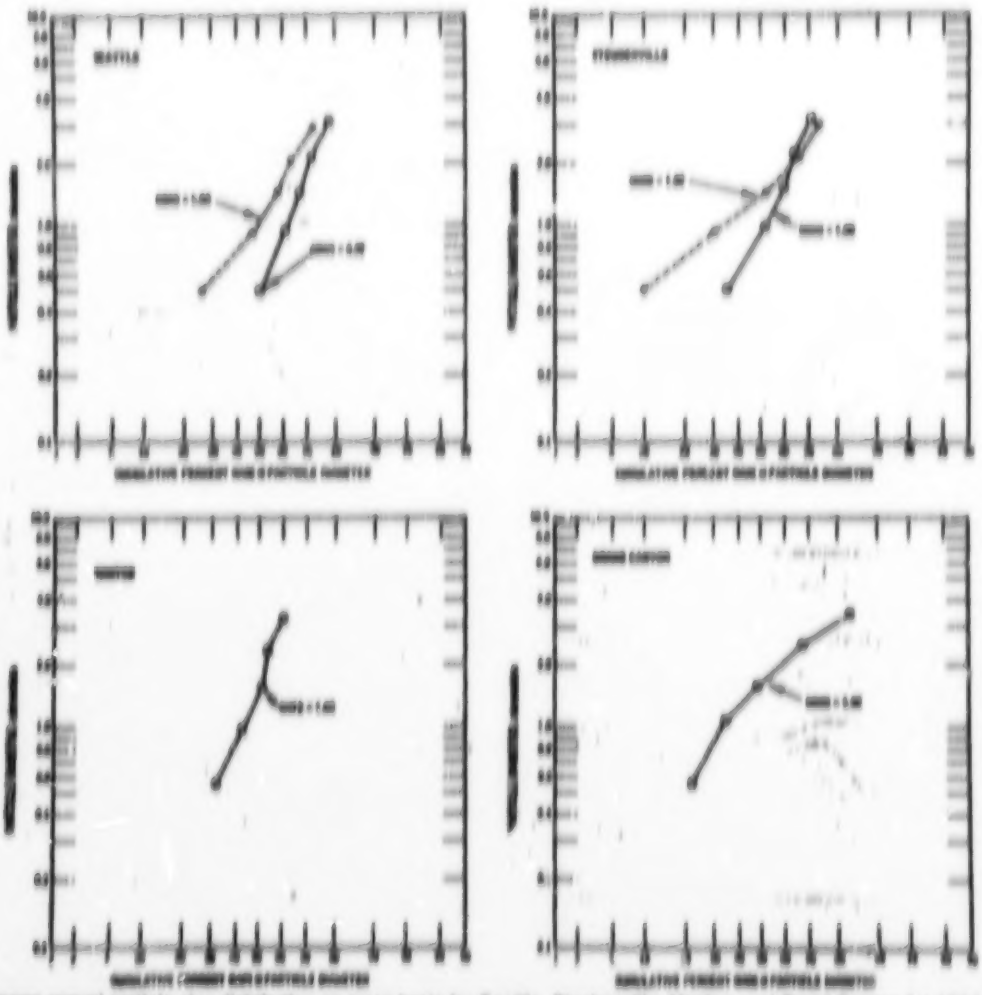

Figure 3. Average annual particle size distribution on mass basis for Seattle, Steubenville, Manteo, and Grand Canyon for 1971 (O — O) and 1972 (● · · · ●)

the other hand, Figure 5 indicates that during the second and third quarters, April through September, the quarterly average MMD generally decreased in size except for some periods at Seattle and Steubenville. Increased photochemical aerosol formation along with a reduction in heating activities during the summer period probably contributed to this downward trend.

The annual trend of an increasing MMD is more difficult to understand; however, several possible explanations may be offered. First, it has been suggested (31) that the impactor jets on the lower stages may become plugged with particulate matter during operation, thereby shifting the distribution erroneously toward a larger median size. However, the impactor stages were cleaned periodically during the sampling period, and inspection of the impaction collection surfaces did not indicate that plugged jets were a problem. Further, in a previous study in Great Britain (19), identical impactors operated intensively over a five-month period provided no evidence of particle growth and exhibited no apparent plugging of the impactor jets. If partial clogging of the jets did occur, it probably did not distort the results to any large extent.

Second, perhaps the most obvious explanation for the increasing MMD trend would be associated with a decrease in the concentration of fine particles resulting from gas to aerosol transformations primarily from stationary emission sources. This explanation seems especially plausible in view of the reduction of gaseous SO_2 which can be oxidized to form particulate SO_4 (38). A decrease in the concentration of fine secondary particles would produce a net shift in the MMD to a larger size. However, examination of the available data summarized in Table III does not provide support for this reasoning. Table III presents the average annual concentration levels of particulate SO_4, NO_3, and NH_4 for many of the urban sites determined from the Hi-Vol sampling network (37). No sharp reduction in the concentration of these components can be seen with the possible exception of Philadelphia. However, it is unlikely that even in Philadelphia, where the MMD increased by a factor of over 3.5 from 1970 to 1971, can changes in the concentration levels of SO_4, NO_3, and NH_4 (associated with predominantly submicrometer particles) account for increases in the MMD.

The most probable but not necessarily the only explanation may be related to the effectiveness of emission control measures which have progressively been applied since 1970 (32, 33). Emission controls on automobiles have reduced the quantity of nonmethane hydrocarbon and carbon monoxide which are associated with the formation of fine, secondary aerosols. The average emission factors decreased for carbon monoxide from 78 g/mi in 1970 to 68 g/mi in 1972, exhaust hydrocarbons from 7.8 g/mi in 1970 to 6.8 g/mi in 1972, and crankcase and evaporation hydrocarbons from 3.9 g/mi in 1970 to 3.0 g/mi in 1972 (36). Nitrogen oxides, however, showed no decrease in the average emission factor over this period, a finding which is supported in part by the results in Table III.

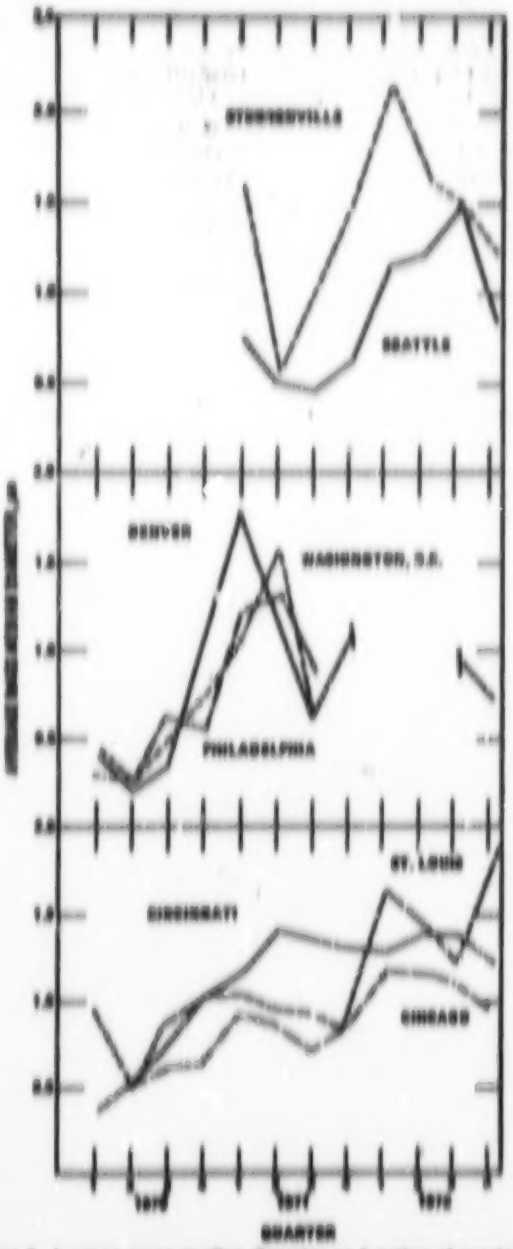

Figure 5. Average mass median diameter as function of quarter for Cincinnati, St. Louis, Chicago, Denver, Washington, D.C., Philadelphia, Seattle, and Steubenville

Table III. Average Annual Concentration of Chemical Components (μg/m³) Derived from NAMN Hi-Vol Sampling Network (37)

	1970	1971	1972
Chicago			
SO_4	14.8	16.1	17.4
NO_3	3.7	4.4	4.4
NH_4	1.1	0.9	0.8
Cincinnati			
SO_4	12.4	11.8	11.9
NO_3	3.5	3.7	3.7
NH_4	0.3	0.4	0.3
Philadelphia			
SO_4	21.9	15.2	16.1
NO_3	5.6	3.8	3.8
NH_4	2.1	0.7	0.6
Denver			
SO_4	4.5	5.0	6.6
NO_3	3.1	3.1	3.6
NH_4	0.1	0.0	0.1
St. Louis			
SO_4	a	12.5	16.3
NO_3	a	2.7	3.9
NH_4	a	0.1	0.2

a Insufficient data available.

Other factors may have a bearing on the apparent increase in median particle size including the effect of stationary control devices in removing coarse particles, growth of fine particles through coagulation processes (39), and the increase in size of hygroscopic particles by humidity. Little data are available, however, to assess the effect of these mechanisms on aerosol growth processes. Further studies need to be made to verify these results, to determine whether the trend of increasing particle size is continuing, and to define those effects which are associated with control measures.

It is encouraging, however, that many urban areas appear to be experiencing not only a decrease in the mass concentration of airborne particulate matter but also an increase in the aerodynamic median size, thereby reducing the inhalation threat. A complete assessment of the health hazard from inhaled particles cannot be made, of course, without characterizing the chemical composition of airborne particulate matter as a function of particle size.

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